One hundred years ago, Heike Kamerlingh Onnes first liquefied helium in a cryogenic laboratory whose excellence and scale were unparalleled. Creating, staffing, and running the Leiden laboratory required more than just scientific skill.

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On the day

Heike Kamerlingh Onnes first liquefied helium, his laboratory at Leiden University briefly encompassed the coldest place on Earth. It was a wet and windy 10 July 1908. Kamerlingh Onnes, awakened before dawn by his wife Betsy, ordered a carriage at 5am to take him from his fine country house on the river Galgewater to his laboratory in the center of the Dutch university town.

His technicians were already hard at work. The day before, they had increased the stock of liquid air to 75 liters, checked the apparatus for leaks, pumped it out, and then filled it with pure hydrogen. Now the first task was to liquefy the hydrogen. While the pumps thundered away in lab Aa, Kamerlingh Onnes and his assistants rushed around, turning valves, connecting and disconnecting gas cylinders, and carefully watching pressure gauges and thermometers. They didn’t stop for lunch. By 1:30pm, 20 liters of liquid hydrogen had been tapped into Dewar flasks, enough to launch the attack on helium in the adjoining lab E’.

Johannes Diderik van der Waals’s law of corresponding states provided a generalized equation of state for nonideal gases scaled to the critical parameters of a particular gas. It told Kamerlingh Onnes how much hydrogen he would need and how much time the helium experiment would take. “These remained just below the limit at which one would have to advise against doing the experiment in the way designed,” he later wrote in his report for the Royal Netherlands Academy of Arts and Sciences. “But just how close we came to that limit only became clear afterwards.” In short, success balanced on a knife-edge.

While the hydrogen liquefier faithfully turned out its 4 liters per hour, a pump throbbed away, maintaining a vacuum in the helium apparatus and removing impurities from the helium gas. At 2:30pm it was time to start cooling the helium. Only 30 minutes later, the temperature had already fallen to 93 K. At 4:20, after a worried Betsy had come to see how the experiment was going commented that the thermometer appeared to be standing in a bath of liquid. Could he be right? When Kamerlingh Onnes pointed a lamp obliquely through the Dewar flasks of liquid air and liquid hydrogen at the glass containing the helium, he could indeed make out a liquid surface! The electric wires of the thermometer were clearly poking through it. “After the surface had once been seen,” he wrote in his report, “it was not lost sight of again. It stood out sharply defined like the edge of a knife against the glass wall.”

Kamerlingh Onnes versus Dewar

The experiment had produced 60 ml of liquid helium, just enough for a little teacup. But it was big science—big by the
standards of the early 20th century—that paved the way. It’s useful to compare the strategies and technical resources of the two main combatants in this cold war on liquid helium, the last of what had been called the permanent gases—namely, those gases that could not be liquefied by compression at then accessible temperatures. The contenders were Heike Kamerlingh Onnes of Leiden University and James Dewar of the Royal Institution in London.

Kamerlingh Onnes, born in 1853, inherited a great deal from his father besides weak lungs. Both loved hard work, and both were manufacturers. While Harm, the father, produced roof tiles, his son Heike produced cold. He was appointed professor of experimental physics at Leiden in 1882, Heike had set up a research program to test and develop the molecular theories of his friend and fellow-countryman van der Waals, in particular the equation of state for a nonideal gas and the law of corresponding states.

The gases to be studied had to be simple, preferably containing no more than two atoms per molecule—for example nitrogen, hydrogen, oxygen, helium, and argon. Such gases condense only at extremely low temperatures. So Kamerlingh Onnes found himself obliged to create a cryogenic laboratory. In 1893 the famous Leiden cascade, consisting of three cooling cycles, started to produce liquid oxygen in quantity. In such cascade processes, pioneered by Swiss physicist Raoul Pictet, sequential stages of different circulating gases are arranged in order of decreasing condensation temperature—that is, in order of increasing volatility. The least volatile gas is cooled and condensed first. Its evaporating vapor is then used to cool and condense the next, more volatile gas. The cascade continues down to the condensation temperature of the most volatile gas in the sequence.

The Leiden cascade, by itself, could not liquefy hydrogen. That’s essentially because the freezing point of oxygen (54 K) exceeds hydrogen’s 33-K critical temperature—that is, the temperature above which gas and liquid are no longer distinct phases. So liquefying hydrogen, whose 20-K boiling temperature is lower than that of any other gas but helium, required iterative Joule–Thomson cooling. But the cascade did provide a continuous supply of liquid air for precooling the hydrogen gas in the laboratory’s indispensable hydrogen liquefier, which began operation in 1906.

James Dewar (1842–1923) grew up in Kincardine-on-Forth, Scotland, where his father ran a pub and a wine business. In the winter of 1852, young James fell through the ice on a pond and became seriously ill with rheumatic fever. During his long convalescence, a local furniture-maker taught him to build violins to develop the muscles in his fingers and arms—an activity to which Dewar later ascribed his dexterity in the laboratory. He studied at the University of Edinburgh and in 1875 secured a chair in experimental physics at Cambridge.

In that bastion of academic tradition, the stocky, opinionated, and irascible Scotsman soon acquired a name for bad lectures and coarse language. In 1887 he succeeded John Tyn dall as director of the Royal Institution in London. By then Dewar had earned his spurs in the science of cold. By means of Joule–Thomson iterative cooling, Dewar succeeded in liquefying hydrogen in 1898. At that point, only one gas still resisted liquefaction: helium.

Dewar was a real showman. He demonstrated his liquefaction of hydrogen to colleagues and the cream of London society at a celebration of the Royal Institution’s 100th anniversary in 1899. The event was immortalized by Henry Jamyn Brooks in a painting that still hangs at the institution.

Dewar’s best-known contribution to cryogenics is the Dewar flask, a double-walled glass container with a vacuum between the walls, silver coated to minimize thermal conduction and radiation. In December 1892 Dewar demonstrated that invention at a Friday evening lecture. With great theatricality, he smashed the tip of one of his vessels containing liquid oxygen. The vacuum space immediately filled...
with air and the liquid oxygen started boiling furiously. The first attempt to liquefy helium had been in 1895. In March of that year, William Ramsay, professor of chemistry at University College London, announced that he had demonstrated the presence of helium on Earth. That the Sun contained this new element had been known spectroscopically since 1868. On Earth, helium comes from alpha decay of radioactive nuclei. To determine helium's critical temperature and pressure, Ramsay sent a sample to Karol Olszewski, an old university friend who was doing cryogenic research in Krakow, Poland. When Ramsay had discovered argon the previous year, he had likewise sent a sample to Krakow instead of to the Royal Institution. That had to do with a row between the two Scotsmen that dated back to a letter to The Times in which Dewar had expressed doubts concerning the discovery of argon. Ramsay was not pleased.

At the end of 1895, Olszewski hoped to produce liquid helium through a process of rapid free expansion. That attempt was doomed to failure. For producing both liquid helium and the liquid hydrogen that must serve to precool it, there was simply no alternative to the Joule–Thomson effect. In the case of helium, the gap between the lowest temperature attainable with liquid hydrogen and 5.2 K, helium's critical temperature, could only be bridged by the Joule–Thomson cooling.

Three years lost

In 1895 Carl von Linde in Germany and William Hampson in England independently designed refrigerators for liquid air based on the Joule–Thomson effect. That brought the prospect of liquefying hydrogen and helium within sight. Kamerlingh Onnes was determined to compete with Dewar and Olszewski. But before he could even get going, his chances of winning the race for hydrogen were crushed. The Leiden city council had heard that a professor was experimenting with "explosive devices" on the very site of the gunpowder-ship explosion that had devastated the city center in 1807. So Kamerlingh Onnes was forbidden to use his cryogenic laboratory until the necessary permit had been approved. What followed was a long and tedious bureaucratic struggle that caused a three-year delay. But he finally carried the day, partly thanks to letters of support from Dewar and Olszewski.

When cryogenic activities resumed at the Leiden laboratory on 4 June 1898, the race to liquefy hydrogen had just produced a winner: James Dewar. He had read out his "Preliminary Note on the Liquefaction of Hydrogen and Helium" at the regular meeting of the Royal Society in London. Two days earlier, he said, he had collected 20 cc of liquid hydrogen in a double-walled insulated glass flask in the basement laboratory of the Royal Institution. He had hoped to collect more, but after five minutes his installation had become clogged with freezing air in the pipes. He had succeeded, however, in immersing two thin tubes into the liquid hydrogen. One was open at the top, and the air at the level of the liquid hydrogen froze immediately. The second tube was connected to a spherical vessel containing helium gas, and its immersion triggered a process of condensation. "All known gases have now been condensed into liquids," Dewar proudly concluded. That was rather less than the truth: He had not in fact condensed the helium itself but only an impurity it contained.

That the Leiden hydrogen liquefier was not fully operational until 1906 was largely because Kamerlingh Onnes was adamant about the stringent standards it had to meet. After Dewar's achievement in 1898, there was no point in quickly putting together a piece of equipment that could produce a little liquid hydrogen, which would at best be useful for some minor experiments. Leiden had lost that race. What Kamerlingh Onnes wanted now was a liquefier that would produce several liters of liquid hydrogen per hour in a continuous process, with maximum economy.

Kamerlingh Onnes and his staff faced the enormous task of making the necessary preparations: They had to modify the extra vacuum and compression pumps, build the cryogenic liquefier, fine-tune an installation for purifying commercially available hydrogen, improve the cascade, and incorporate a cycle for liquid air. But when they were finished, the result was something to be proud of.

As soon as Kamerlingh Onnes had subduced hydrogen in 1906, he started planning the assault on helium. Dewar had made a renewed attempt in 1901. After his failed attempt of three years earlier, Dewar reverted to a single-step expansion method pioneered by French engineer Louis-Paul Cailletet, who in 1877 had shattered the myth of permanent gases by liquefying oxygen and nitrogen. Cailletet succeeded by cooling the gas in a single free-expansion step after hydraulically compressing it to very high density. Such a single-step cooling method, if it could be made to work, would require far less helium than did the cyclic Joule–Thomson cooling.

Dewar obtained his helium from the hot springs at Bath, in the form of gas (mainly nitrogen) containing a small proportion (0.05%) of helium. He then removed everything except the helium by chemical reaction, cooling condensation, or freezing in a U-shaped tube immersed in liquid hydrogen at 16 K.

Dewar allowed the helium gas, which was compressed to 80 atmospheres, to expand in the Cailletet tube. But he saw nothing—not even a trace of mist. From the fact that helium produced no mist after expanding at 16 K, Dewar concluded that its critical temperature must be below 9 K.

In Krakow, Olszewski's first attempt to liquefy helium in 1895 had also led him to suspect that the boiling point of helium must be lower than 9 K. In 1905 he tried again. In his second attempt, Olszewski started with liquid hydrogen, used a larger Cailletet tube, and increased the pressure on the helium (derived from the mineral thorianite that had just been discovered in Ceylon) to 180 atmospheres. Again, there was nothing to see. He now concluded that helium's boiling point must be below 2 K and suggested that its liquefaction might prove to be impossible.

When it became clear that the helium could not be subduced in a single-shot Cailletet expansion, the obvious solution was to switch to the approach that had worked in producing liquid hydrogen—a liquefier with circulating helium, a regenerator coil, and a porous expansion valve, all of this precooled with liquid hydrogen. In 1901 Dewar calculated that starting with liquid or solid hydrogen, it must be possible to liquefy helium by that iterative Joule–Thomson method, provided that its critical temperature was no lower than 6 to 8 K, corresponding to a boiling point of 4 or 5 K.

Early in June 1905 Kamerlingh Onnes wrote to Dewar, saying that he was now ready to begin the precise determination of the helium isotherms on the pressure–volume plane, an important stage in determining the critical temperature and a job for which the Leiden laboratory had the ideal facilities. He was desperately impatient to obtain a few liters of pure helium gas, and in the absence of any other supplier he turned to Dewar. To reassure his competitor, Kamerlingh Onnes added that it would take him two years to purify Dewar's helium to the desired standard. And weren't the isotherms the ideal route toward finding a reliable critical point, which would in turn answer the pressing question of whether it was actually feasible to liquefy helium using the Joule–Thomson approach?
But, it seems, Kamerlingh Onnes was asking rather too much of Dewar's generosity. So he turned to Ramsay, who proved more amenable. In response to the request, Ramsay wrote that his helium did not contain any neon—a great advantage over Dewar's—and that the air it contained could easily be removed by passing the gas through liquid hydrogen, after which remaining traces of hydrogen could be removed chemically.

In Dewar's laboratory the helium cycle was not going well. Robert Lennox, the chief lab assistant, had wanted to make a metal liquefier. But Dewar, like Kamerlingh Onnes, wanted glass so he could see what was going on. Dewar's installation devoured liquid hydrogen. Six liters of it evaporated within just four minutes of helium circulation. Worse still, the installation he was using to purify the helium from Bath was malfunctioning. As a result, the stopcocks used in the helium cycle became clogged with frozen neon. Collaboration with Ramsay, a past master at purifying inert gases, might have saved the day. But that was impossible after the row back in 1894. Besides, Dewar also needed a pump that could compress the helium without contaminating the cycle. And to crown it all, a young Royal Institution technician turned the wrong stopcock one day, and Dewar's entire stock of pure helium leaked away overnight!

**Sinking hopes and helium snow**

In 1905 in Leiden, a start was made on measuring helium isotherms at temperatures ranging from 373 K down to 14 K, the freezing point of hydrogen. But before the isotherms had been determined, an alternative method of finding critical temperature presented itself. In November 1906 Kamerlingh Onnes submitted a short communication on a spectacular phenomenon: a gas that sinks in liquid. This “barotropic” effect involved helium gas compressed above liquid hydrogen until it sank. Applying van der Waals's theory of binary mixtures to that observation, one would conclude that the critical temperature of helium must be below 2 K. It meant that the Joule–Thomson method held out little prospect of success with helium, even starting with solid hydrogen.

The helium isotherms, which provided the only way of accurately determining critical temperature, now became all the more important. At the end of 1907, Kamerlingh Onnes established that helium's inversion temperature was 23 K and he concluded that its critical temperature was 5.3 K. So Joule–Thomson expansion of helium would indeed lead to liquefaction if the process was started at the freezing point of hydrogen.

Heartened by that result, he decided to try condensing helium using one-step expansion in a glass test tube. There was no harm in trying. On 28 February 1908, he allowed 7 liters of helium starting at 14 K to expand from 100 atmospheres to 1 atmosphere. To his not inconsiderable astonishment, the glass tube beneath the expansion stopcock filled with a flaky, snowlike mass. The most he had hoped for was a faint mist!

The news spread through the laboratory like wildfire:
After the success of 10 July 1908, Kamerlingh Onnes’s cryogenic laboratory possessed a monopoly on liquid helium for the next 15 years. Not until 1923 did John McLennan at the University of Toronto succeed in producing liquid helium. That was after McLennan had gone to look around the Leiden laboratory and had been given the plans for the Leiden hydrogen and helium liquefiers in exchange for the cylinder of helium gas that he brought with him.

How did Leiden succeed in holding on to its unique position for so long? To start with, Kamerlingh Onnes was assisted by an exceptionally gifted team of technicians, headed by Gerrit Flim and Oskar Kesselring. The helium liquefier was a complex instrument, and keeping the circulating helium gas pure called for ingenious pumps such as the Cailletet compressor (shown at right) with modifications introduced by Kamerlingh Onnes.

In 1883, Louis-Paul Cailletet had devised a new type of gas compressor whose distinguishing feature was the addition of a column of mercury above the piston. Because the mercury followed the contours of the cylinder, the pump was extraordinarily efficient. For maintaining the purity of helium, however, Cailletet’s design had a problem. The mercury was in contact with the piston’s glycerin lubricant. After numerous disappointments, Kamerlingh Onnes finally succeeded in reconfiguring the Cailletet compressor so that there was no possible contact between the glycerin contaminant and the gas being compressed. The purity of the circulating helium was a crucial issue. In London, James Dewar constantly struggled with frozen taps and clogged pipes. He had nothing like the Leiden Cailletet compressor.

A still more important factor was the scarcity of helium gas. After William Ramsay’s discovery of the new element, pure helium gas was initially very rare. Isolating it from monazite sand was an elaborate and time-consuming process, for which few laboratories possessed the necessary expertise or resources. In 1909 an alternative source of helium presented itself when Kamerlingh Onnes was visited by Bertram Boltwood, an American radiochemistry pioneer who was working with Ernest Rutherford in Manchester. When he returned to England from Leiden, Boltwood wrote to his friend the chemist H. S. Miner of the Welsbach Light Co. in New Jersey about Kamerlingh Onnes’s need for more helium gas.

The Welsbach firm produced thorianite mantles for gas lamps. Carl Auer von Welsbach, the company’s founder, had applied for a patent in 1889 for mantles to which thorium oxide was added. The company would be processing large quantities of the thorianite, a mineral that had been discovered in 1904. It’s mostly ThO₂, and it contains helium from the alpha decay of thorium. Welsbach was willing to send the helium to Kamerlingh Onnes, free of charge, in return for the monazite sand waste (which still contained thorium) being discarded by the Leiden laboratory.

In 1911 Welsbach sent him eight steel gas cylinders that yielded a total of 125 liters of pure helium gas. A second consignment arrived in March 1913, after urgent entreaties from the Leiden laboratory, which had lost 80 liters of helium due to the breakage of a glass container. The hoped-for third consignment never came. After the outbreak of World War I, helium was found to be of military value for airships.

After the war, when Kamerlingh Onnes was desperate to replenish his dwindling stocks, the US Bureau of Mines came to the rescue. In the summer of 1919, the bureau’s Frederick Cottrell visited the Leiden laboratory. He explained that a factory in Texas was extracting helium from natural gas. It had some 4 million liters of helium ready to be shipped to France when the armistice ended the war. Kamerlingh Onnes promptly telegraphed an American admiral who was involved in designing airships: “According advice American colleagues I pray to send helium for continuing experiments lowet temperatures.” It worked. In the fall of 1919, five cylinders arrived in Leiden, yielding hundreds of liters of 96%-pure helium.

The Bureau of Mines decided that since it was so ampley supplied with helium, it should set up its own cryogenic laboratory, including a helium liquefier. But the plans came to nothing. The first liquefier in the US eventually materialized in 1931 at the National Bureau of Standards in Washington, DC. By 1925 physicist Walther Meissner in Berlin also possessed liquid helium. Once Oxford, Cambridge, and Kharkov in the Soviet Union had followed suit around 1930, the proliferation of liquid helium was worldwide.
“Solid helium!” He announced it the very next day at an academy meeting, and he sent Dewar a telegram. Six weeks later the solid helium was clearly seen to have been a mistake. The observed phenomenon was in fact attributable to the presence of hydrogen in the helium. The hydrogen had frozen during the expansion and then rapidly evaporated.

So now Kamerlingh Onnes decided to build a helium liquefier with a regenerator coil and Dewar flask, a miniature of his successful hydrogen liquefier. The downscaling was guided by van der Waals’s law of corresponding states. After three months of working flat out, the group had completed the apparatus assembly and there was enough pure helium to attempt liquefaction.

Kamerlingh Onnes had obtained the extra helium (a few hundred liters) required for the Joule–Thomson circulation method from his brother Onno, who ran a commercial information office in Amsterdam. Onno arranged the delivery of two sacks of helium-containing monazite sand that had been shipped to Hamburg from North Carolina. Purifying the helium gas was a particularly grueling task. It demanded the concentrated efforts of four chemists for several months.

They heated the sand grains, causing them to explode and release the gas. To remove impurities, they then cooled the gas with liquid air, burned it with oxygen over copper oxide, compressed it at the temperature of liquid hydrogen, and passed it over a charcoal filter. At last, on 10 July, the helium gas was finally pronounced sufficiently pure.

**An astonishing achievement**

The liquefaction of helium marked the culmination of Kamerlingh Onnes’s career as a scientist. He was almost 55 years old. Given the apparatus he was using, with high-pressure pipes and a regeneration coil, the fact that he succeeded with only 200 liters of helium—and 160 liters in reserve—was an outstanding achievement.

At the time, he regretted that he wasn’t able to use the laboratory’s large Burckhardt vacuum pump in his first liquefaction of helium. It could have pumped down to a pressure of 2 mm of mercury. So the historic experiment had to make do with a vacuum pressure that went no lower than 0.03 atmosphere. Therefore he could only get the helium down to 1.7 K, insufficient—he thought—to freeze it. It later became clear that freezing helium actually required high pressure.

The purity of the helium used in the historic experiment was beyond dispute. Stopcocks continued to function perfectly, and even the final remains of the helium fluid were crystal clear. The measured boiling point, as determined with a helium gas thermometer, was corrected to 4.5 K, a few tenths of a degree too high. Critical pressure and density were both surprisingly low. Kamerlingh Onnes was also struck by the volatility of liquid helium. He had in fact cooled the helium below the lambda point (2.17 K), at which we now know it enters its superfluid phase.

The news of the liquefaction of helium was a blow for Dewar. He and Lennox soon fell out. Lennox, who had lost an eye in one of the all-too-frequent mishaps in the cryogenic laboratory, resigned and swore never again to set foot in the Royal Institution as long as Dewar lived. And he remained true to his word.

After Kamerlingh Onnes’s victory in the race for liquid helium, Dewar said that two problems had plagued him: obtaining a sufficient quantity of pure helium, and making enough liquid hydrogen to keep the helium cycle operational (see the box on page 40). All that, he admitted, called for an industrial approach, quite alien to tradition at the Royal Institution. “In my work I have never been able to do anything unless substantially with my own hands,” Dewar had written to Kamerlingh Onnes in 1904. “In pioneering work, assistants are a waste.”

Kamerlingh Onnes, by contrast, had assembled a small army of “blue-collar boys” around him, pupils of the School of Instrument Makers attached to the Leiden physics laboratory. This unique school, founded by him and fully integrated into his laboratory, was headed by outstanding technicians such as the brilliant instrument maker Gerrit Flim and the master glass blower Oskar Kesselring.

Kamerlingh Onnes’s entrepreneurial abilities proved invaluable. He built up a cryogenic laboratory of international status. Its size and staff were unequalled anywhere in the
world. That called for more than a great talent for physics. Visiting the Leiden cryogenic laboratory with its profusion of tubes, faucets, gas holders, liquefiers, Dewar flasks, cryostats, workshops, instruments, clattering pumps, and droning engines would have felt like entering a factory.

It was indeed a “cold factory,” with Professor Kamerlingh Onnes as its director, determining policy and exercising tight overall control. As the director of such an enterprise, he also set up a well-run organization presided over by an administrative supervisor, a research team that included assistants and graduate students, a manager, instrument makers, glass blowers, laboratory assistants, technicians, an engineer, and an assistant supervisor. There was also a small army of trainee instrument makers to perform any number of odd jobs.

This big-science approach, novel in its combination of focus and scale, could only succeed with someone at the helm who had persistence, courage, willpower, vision, and indestructible patience. Someone who ruled with a firm hand, but who at the same time had a gift for winning people over, persuading them, and securing their loyalty.

The enterprise also needed someone with a peerless ability to manipulate the powers that be. Kamerlingh Onnes constantly warned that what had taken years to achieve in his laboratory was in danger of being destroyed, and he kept up his dire warnings until the authorities gave their “expensive professor” the space and resources he needed. He was also a brilliant networker, with a keen eye for useful contacts both within and beyond the field of physics. He pampered his guests and was far too shrewd to quarrel—even with Dewar—or to make enemies who might harm his interests.

In short, Kamerlingh Onnes brimmed with organizational and social instinct, without which his mission in Leiden would have had little chance of success. He was a sound scientist, but his cryogenic laboratory owed its success to his talent for organization, his social skills, and his unswerving focus on extremely low temperatures.

From 10 July 2008 until 10 May 2009, the Boerhaave Museum in Leiden will present the exhibition “The Quest for Absolute Zero.”

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